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Low-temperature synthesis and microwave dielectric properties of trirutile-structure $MgTa_2O_6$ ceramics by aqueous sol–gel process

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Abstract

Trirutile-structure $MgTa_2O_6$ ceramics were prepared by aqueous sol–gel method and microwave dielectric properties were investigated. Highly reactive nanosized $MgTa_2O_6$ powders were successfully synthesized at 500 °C in oxygen atmosphere with particle sizes of 20–40 nm. The evolution of phase formation was detected by DTA–TG and XRD. Sintering characteristic and microwave dielectric properties of $MgTa_2O_6$ ceramics were studied at different temperatures ranging from 1100 to 1300 °C. With the increase of sintering temperature, density, ε_r and $Q \cdot f$ values increased and saturated at 1200 °C with excellent microwave properties of $\varepsilon_r \sim 30.1$, $Q \cdot f \sim 57{,}300$ GHz and $\tau_f \sim 29$ ppm/°C. The sintering temperature of $MgTa_2O_6$ ceramics was significantly reduced by aqueous sol–gel process compared to conventional solid-state method.
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Keywords: A. Sol–gel process; $MgTa_2O_6$; Nanopowder synthesis; Microwave dielectric properties

1. Introduction

The rapid progress in mobile and satellite communication system were creating high demands for the development of microwave dielectric materials with a high quality factor, an appropriate dielectric constant, and a near-zero temperature coefficient of resonant frequency. High quality AB_2O_6 (A = Ca, Mg, Mn, Co, Ni, Zn and B = Nb, Ta) compounds were investigated as microwave dielectric resonators by Lee et al. [1]. Lee et al. reported that $MgTa_2O_6$ (MT) sintered at 1550 °C exhibited relative dielectric constant (ε_r) of 30.3, quality factor ($Q \cdot f$) of 59,600 GHz, and temperature coefficient of resonant frequency (τ_f) of 30 ppm/°C by solid state process. Obviously high sintering temperatures would limit their applications for practical cases, so the reduction of sintering temperatures was desirable to enable commercial applications such as in integrated circuits. Usually it was believed that lowering sintering temperature could be achieved by many methods such as adding glass flux and using starting materials with smaller particle sizes. As we known, adding glass flux usually caused detrimental effect on microwave properties of ceramics. The synthesis of MT powders was widely investigated [2–4] and now in order to reduce sintering temperature and improve sintering ability there were many other investigations of chemical processing or special methods [5,6], which were developed as alternatives to the conventional solid-state reaction of mixed oxides for producing ceramics using starting materials with smaller particle sizes. Among these wet chemical techniques, the sol–gel was undoubtedly one of useful process for producing powders with good control over stoichiometry and homogeneity, yielding nano-sized particles and widely used in many other ceramics system [7–10]. However, few researches about microwave properties of MT ceramics fabricated by aqueous sol–gel process were reported in the present literatures.

In this paper the sol–gel method was used to synthesize nano-sized powders as precursors for preparation of MT ceramics instead of other methods. The whole process involved all complexation of aqueous metal ions by non-toxic poly functional carboxyl and avoided complex steps such as refluxing of alkoxides, resulting in less time consumption compared to other techniques. The evolution of MT phase

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formation and microwave dielectric properties of MT ceramics as a function of sintering temperatures were investigated in detail. Experimental results showed that the preparation of MT ceramics with retaining excellent microwave properties could be obtained efficiently and simply at low sintering temperatures by aqueous sol–gel process.

2. Experimental

Analytical-grade Ta_2O_5, K_2CO_3, $Mg(NO_3)\cdot 6H_2O$, HNO_3, citric acid (CA) and ethylene glycol (EG) were used as raw materials to synthesize MT nanopowders as shown in Fig. 1. Firstly, the mixture of Ta_2O_5 and K_2CO_3 was co-melted at 900 °C in order to obtain K_3TaO_4 compounds according to phase diagrams. Subsequently, K_3TaO_4 compounds were dissolved in distilled water, then pH value was controlled at ~2 to ensure the formation of $Ta(OH)_5$ precipitate. The whole formation process of $Ta(OH)_5$ phases could be formulated from Eqs. (1)–(4). Thirdly, $Ta(OH)_5$ precipitate was filtered off and washed with distilled water for six times to remove K^+ ions and then dissolved completely in citric acid water solution by continuous magnetic stirring at 300 rpm for 15 min. Meanwhile, a stoichiometric amount of $Mg(NO_3)\cdot 6H_2O$ was added to above solution and then the solution was stirred for another 30 min. Finally, ethyl alcohol (20–40 ml) was added to the as-prepared mixed solution in drops and stirred for 1 h to form a transparent and stable sol. pH of the solution was maintained in the range of 3.5–5 by adding buffering agents. The sol was heated at 80–90 °C for 1 h to obtain a xerogel. The xerogel was decomposed at 500 °C using a muffle furnace for crystallization in oxygen atmosphere. The as-prepared powers were ball milled in a polyethylene jar for 4 h using ZrO_2 balls in ethanol medium to reduce conglobation phenomena. Powders were then mixed with polyvinyl alcohol as a binder, granulated and pressed into cylindrical disks of 10 mm diameter and about 5 mm height at a pressure of about 200 MPa. These pellets were preheated at 600 °C for 4 h to expel the binder and then sintered at selected temperatures for 2 h in air at a heating rate of 5 °C/min.

$$3K_2CO_3 + Ta_2O_5 \rightarrow 5K_3TaO_4 + CO_2. \quad (1)$$

$$6K_3TaO_4 + 5H_2O \rightarrow 18K^+ + Ta_6O_{19}^{8-} + 10OH^-. \quad (2)$$

$$4K^+ + Ta_6O_{19}^{8-} + 4OH^- + 8H^+ \rightarrow K_4H_4Ta_6O_{19}\downarrow + 4H_2O. \quad (3)$$

$$K_4H_4Ta_6O_{19} + 15H^+ + 11OH^- \rightarrow 6Ta(OH)_5\downarrow + 4K^+. \quad (4)$$

In order to analyze the evolution of MT phase formation, the as-formed MT xerogel was characterized using thermogravimetry (TG) and differential thermal analysis (DTA) to study its thermal properties. Phase analysis of MT powders was conducted with the help of a Rigaku diffractometer (Model D/MAX-B, Rigaku Co., Japan) using Ni filtered Cu Kα radiation (λ = 0.1542 nm) at 40 kV and 40 mA settings. Based on XRD analysis, raw MT powders were examined for their morphology and particle size using a transmission electron microscopy (Model JEOL JEM-2010, FEI Co., Japan). Bulk densities of sintered ceramics were measured by the Archimedes method. An HP8720ES network analyzer (Hewlett-Packard, Santa Rosa, CA) was used for the measurement of microwave dielectric properties. Dielectric constants were measured using Hakki-Coleman post-resonator method by exciting the TE011 resonant mode of dielectric resonator using an electric probe as suggested by Hakki and Coleman and Courtney [11]. Unloaded quality factors were measured using TE01d mode by the cavity method [12]. All measurements were made at room temperature and in the frequency of 8–10 GHz. Temperature coefficients of resonant frequency were measured in the temperature range of 25–85 °C.

3. Results and discussion

Fig. 2 showed TG–DTA curves of MT xerogel in pure oxygen atmosphere at a heating rate of 10 °C/min. The results indicated that obvious weight losses began at 250 °C and all chemical reactions involving weight losses, such as decom-

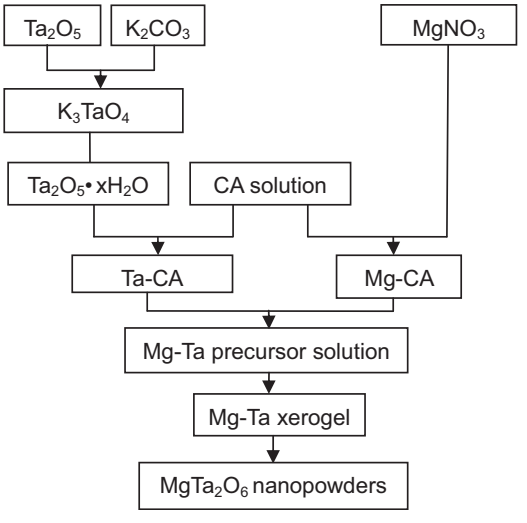

Fig. 1. Chart for the synthesis of MT nanopowders by aqueous sol–gel processing.

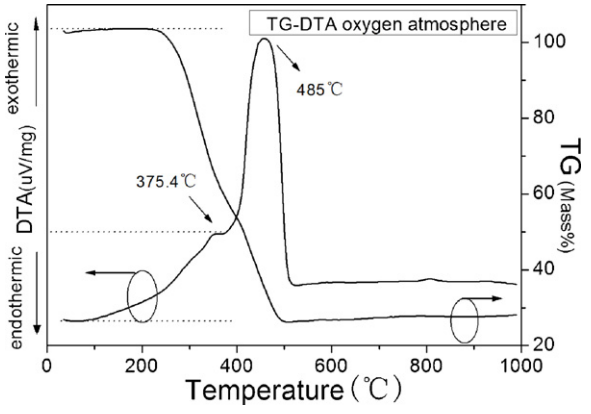

Fig. 2. TG–DTA curves of MT xerogel in oxygen atmosphere.

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